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Arbitrary Lagrangian–Eulerian finite-element method for computation of two-phase flows with soluble surfactants

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ABSTRACT

A finite-element scheme based on a coupled arbitrary Lagrangian–Eulerian and Lagrangian approach is developed for the computation of interface flows with soluble surfactants. The numerical scheme is designed to solve the time-dependent Navier–Stokes equations and an evolution equation for the surfactant concentration in the bulk phase, and simultaneously, an evolution equation for the surfactant concentration on the interface. Second-order isoparametric finite elements on moving meshes and second-order isoparametric surface finite elements or solve these equations. The interface-resolved moving meshes allow the accurate incorporation of surface forces, Marangoni forces and jumps in the material parameters. The lower-dimensional finite-element meshes for solving the surface evolution equation are part of the interface-resolved moving meshes. The numerical scheme is validated for problems with known analytical solutions. A number of computations to study the influence of the surfactants in 3D-axisymmetric rising bubbles have been performed. The proposed scheme shows excellent conservation of fluid mass and of the total mass of the surfactant.

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1. Introduction

The presence of surface active agents (surfactants) significantly alters the dynamics of multiphase flows. Surfactants lower the surface tension on the interface and since, in general, their concentration along the interface is not uniform, Marangoni forces are induced. These properties of surfactants offer the possibility of controlling the dynamics of multiphase flow systems.

Surfactant-controlled multiphase flow systems are widely used in scientific, engineering and biomedical applications. For example, surfactants can be used to manipulate very small droplets and bubbles [6,11] which is useful in flow-focusing devices [2,29]. The presence of surfactants in pulmonary alveoli is essential for the proper functioning of the defense mechanism of lungs [9,17,25]. A lack of pulmonary surfactants in premature neonates causes the respiratory distress syndrome (RDS) [3].

A mathematical model describing interface flows with soluble surfactants consists of the time-dependent Navier–Stokes equations coupled with the bulk and the surface evolution equations for the concentration of surfactants in the bulk fluid phase and on the interface, respectively. Since the interface has to be captured/tracked during the computations, the solution of the surface evolution equation on the deforming interface is one of the main challenges in the computation of flows with

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surfactants. The exchange of surfactants between the interface and the bulk phase increases the complexity further. The force balance along the interface couples the surfactant concentration on the interface with the dynamics of the flow field and makes the computation even more challenging. Thus, a realistic study of the effect of surfactants in interface flows depends largely on the accuracy of the numerical scheme used.

Most of the previous numerical studies on the effect of surfactants in interface flows are confined to flows with insoluble surfactant, where the surfactant mass transfer between the interface and the bulk phase is neglected. For interface flows with insoluble surfactants a number of numerical schemes based on popular interface capturing/tracking methods such as volume-of-fluid [19,27], level set [33] and arbitrary Lagrangian–Eulerian method [14,26] have been proposed by several authors. Boundary integral methods [4,11,23] and immersed boundary methods [21,22] have also been used to study the effect of insoluble surfactants in interface flows. The complexity of computations increases when the surfactant mass transfer between the interface and the bulk phase is taken into consideration. In particular, the conservation of the total mass of surfactants is a challenge in the computation of interface flows with soluble surfactants. A narrow transition layer in which the concentration of the bulk surfactants varies rapidly will occur adjacently to the interface when the bulk Peclet number is large [5]. It is well known that standard Galerkin finite-element discretizations applied to convection-dominated convection-diffusion equations induce spurious oscillations in the numerical solution unless the mesh size is small enough to capture the transition layer. In this case, stabilization methods [28] like the streamline-upwind Petrov–Galerkin (SUPG) method [8] or the local projection stabilization method [15] could be applied to stabilize the bulk concentration equation. Alternatively, layer-adapted meshes [28] near the interface combined with standard discretizations can be used. In the considered test examples, we followed the second way and have chosen the mesh very fine near the interface. Note that the ALE approach produces a convection term with a relative velocity vanishing on the interface. This might be the reason that in our computations no stabilization was needed. However, a complete discussion of this topic is beyond the scope of the paper.

Recently, a front-tracking method has been developed for computations of interface flows with soluble surfactants [34]. In the front-tracking method, the interface is represented by connected Lagrangian marker points, which move with the local fluid velocity interpolated from the background stationary Eulerian grid. In [34], the adsorption and desorption balance condition for the surfactant mass transfer between the interface and the bulk phase has been incorporated at the interface, which is constructed from the Lagrangian marker points. Unfortunately, this method does not conserve the total surfactant mass. Another variant of the front-tracking method for interface flows with soluble surfactants has been proposed in [24]. With the assumption that the surfactant mass transfer occurs within a thin adsorption layer adjacent to the interface, a better conservation of the total mass of surfactants has been achieved. Also, a meshfree smoothed-particle hydrodynamics (SPH) method has been proposed in [1] for two-dimensional Stokes interface flows with soluble surfactants. In the SPH method, Lagrangian particles are marked with a color function to distinguish different fluid phases. The authors incorporated surface and Marangoni forces into the SPH model using the continuum surface force (CSF) technique [7]. Further, the surface evolution-equation has been approximated in a transition layer adjacent to the interface in [1].

In this paper, we present an accurate and efficient sharp interface numerical method based on a coupled arbitrary Lagrangian–Eulerian and Lagrangian approach. Since the interface is resolved by this method, surface forces, Marangoni forces and jumps in the material parameters can be accurately incorporated into the model. As shown in [12] spurious velocities can be successfully suppressed using isoparametric finite elements, discontinuous pressure approximations, and the Laplace–Beltrami technique for representing the curvature. Furthermore, no separate lower-dimensional interface mesh is needed for approximating the surface evolution-equation; instead we use the discrete representation of the interface directly. In addition, the incorporation of the adsorption and desorption balance condition for the surfactant mass transfer relation is straightforward in the sharp interface method considered.

The paper is organized as follows. In Section 2, the governing equations and the transformation into their dimensionless form are presented. The finite-element scheme based on the coupled arbitrary Lagrangian–Eulerian and Lagrangian method for interface flows with soluble surfactants is described in Section 3. The accuracy of the proposed numerical scheme is validated for test examples with known analytical solutions in Section 4. Furthermore, numerical results for a rising bubble are given. Finally, in Section 5 we summarize the results.

2. Governing equations

We consider an incompressible two-phase flow with a soluble surfactant in a bounded domain $\Omega \subset \mathbb{R}^3$ with Lipschitz continuous boundary $\partial \Omega$. We assume that a liquid droplet filling $\Omega_1(t)$ is completely surrounded by another liquid filling $\Omega_2(t) = \Omega \setminus \overline{\Omega}_1(t)$ and the liquids are immiscible. Here, *t* is the time in a given time interval [0, T]. The interface between the liquids is denoted by $\partial \Omega_F(t) := \partial \Omega_1(t) \cap \partial \Omega_2(t)$. The concentration of the soluble surfactants on the interface $\partial \Omega_F(t)$ influences the surface tension and thus the dynamics of the flow.

2.1. Navier–Stokes equations

10

The two-phase flow in Ω is described by the time-dependent incompressible Navier–Stokes equations

$$\rho_k \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) - \nabla \cdot (\mathbb{T}_k(\mathbf{u}, p)) = \rho_k g \mathbf{e}, \quad \nabla \cdot \mathbf{u} = 0, \quad \text{in } \Omega_k(t) \times (0, T]$$
(1)

for k = 1, 2. The Navier–Stokes equations are completed by the initial condition,

$$\Omega(\mathbf{0}) = \Omega_0, \quad \mathbf{u}|_{t=0} = \mathbf{u}_0,$$

the kinematic and force balancing conditions

$$\mathbf{w} \cdot \mathbf{v} = \mathbf{u} \cdot \mathbf{v}, \quad [\mathbf{u}] = \mathbf{0}, \quad \mathbf{v} \cdot [\mathbb{T}(\mathbf{u}, p)] \cdot \mathbf{v} = \sigma(\Gamma)\mathcal{K}, \quad \tau_i \cdot [\mathbb{T}(\mathbf{u}, p)] \cdot \mathbf{v} = \tau_i \cdot \nabla \sigma(\Gamma), \quad \text{on } \partial \Omega_F(t)$$

for *i* = 1, 2. Further, we assume that the boundary $\partial \Omega$ of Ω is fixed in time, and impose homogeneous Dirichlet boundary conditions on it. For the Newtonian incompressible fluid, the stress tensor $\mathbb{T}_k(\mathbf{u}, p)$ and the velocity deformation tensor $\mathbb{D}(\mathbf{u})$ are given by

$$\mathbb{T}_k(\mathbf{u},p) = 2\mu_k \mathbb{D}(\mathbf{u}) - p\mathbb{I}, \quad \mathbb{D}(\mathbf{u}) = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right), \quad i,j = 1, \dots, 3.$$

Here, $\mathbf{u} = (u_1, u_2, u_3)$ is the fluid velocity, p is the pressure, ρ_k and μ_k are the density and the dynamic viscosity of the respective fluid phases, \mathbf{w} on $\partial \Omega_F(t)$ is the interface velocity and \mathcal{K} is the sum of the principal curvatures. Further, \mathbf{v} and τ_i , i = 1, 2 denote the unit outward normal and tangential vectors on $\partial \Omega_F(t)$, \mathbf{e} an unit vector in the opposite direction of the gravitational force, I the identity tensor, and [·] denotes the jump across the interface $\partial \Omega_F(t)$. Further, Γ denotes the surfactant concentration on the interface and $\sigma(\Gamma)$ the surface tension coefficient depending on Γ . In our model, we consider both the linear and nonlinear equation of states for the surfactant and the surface tension relation. Henry's linear equation of states (LEOS)

$$\sigma(\Gamma) = \sigma_1 + RT_a(\Gamma_1 - \Gamma) \tag{2}$$

can be used when the variation of the surfactant concentration around a reference surfactant concentration, Γ_1 , is small, see for example [19,26]. Here, σ_1 corresponds to the surface tension in the reference phase, R is the ideal gas constant and T_a is the absolute temperature. The nonlinear Langmuir equation of state (NLEOS) is given by

$$\sigma(\Gamma) = \sigma_0 + RT_a \Gamma_\infty \ln(1 - \Gamma/\Gamma_\infty), \tag{3}$$

where σ_0 is the surface tension coefficient of the surfactant-free (clean) liquid and Γ_{∞} is the maximum surface packing surfactant concentration. Note that due to the log singularity the NLEOS produces negative values of surface tension when $\Gamma \rightarrow \Gamma_{\infty}$. In some papers, this effect is taken into consideration by introducing a cut-off value for the minimum surface tension [24]. However, in the case of high surface concentrations we believe that a more detailed model is needed to account for the tendency of soluble surfactants to form micelles in the bulk. Nevertheless, we always checked the size of the surface tension reduction to avoid unphysical regimes in our computations.

2.2. Soluble surfactant transport equations

In the present study, it is assumed that the surfactant is soluble only in the outer fluid phase $\Omega_2(t)$. The transport of the surfactant concentration C in $\Omega_2(t)$ is described by the scalar convection–diffusion equation

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C = \nabla \cdot D_c \nabla C \quad \text{in } \Omega_2(t) \times (0, \mathrm{T}].$$
(4)

Eq. (4) is completed with the initial and boundary conditions:

$$C|_{t=0} = C_0 \qquad \text{in } \Omega_2(0),$$

$$v \cdot D_c \nabla C = -S(\Gamma, C) \qquad \text{on } \partial \Omega_F(t),$$

$$v \cdot D_c \nabla C = 0 \qquad \text{on } \partial \Omega.$$

Here, D_c is the diffusive coefficient of the outer phase surfactant concentration C. The source term $S(\Gamma, C)$ is given by

$$S(\Gamma, C) = K_a C(\Gamma_\infty - \Gamma) - K_d \Gamma, \tag{5}$$

where K_a and K_d are adsorption and desorption coefficients, respectively. The surfactant concentration Γ on the interface $\partial \Omega_F(t)$ is described by the surface convection–diffusion equation [30,32]

$$\frac{\partial \Gamma}{\partial t} + U \cdot \underline{\nabla}\Gamma + \Gamma \underline{\nabla} \cdot \mathbf{u} = \underline{\nabla} \cdot (D_s \underline{\nabla}\Gamma) + S(\Gamma, C)$$
(6)

for a given initial concentration $\Gamma|_{t=0} = \Gamma_0$. Here, D_s is the diffusive coefficient of Γ and $U = (\mathbf{u} - (\mathbf{u} \cdot \mathbf{v}) \cdot \mathbf{v})$ is the velocity along the interface. The surface (tangential) gradient operator ∇ is defined by

$$\underline{\nabla}\Gamma := \nabla\Gamma - (\boldsymbol{v}\cdot\nabla\Gamma)\boldsymbol{v}.$$

In this definition, we have assumed that Γ is defined not only on $\partial \Omega_F(t)$ but also in a certain neighborhood. However, it is well known that the restriction of $\underline{\nabla}\Gamma$ on $\partial \Omega_F(t)$ depends only on values of Γ on $\partial \Omega_F(t)$. In Eq. (6), $\underline{\nabla} \cdot \mathbf{u}$ is the tangential divergence of the fluid velocity, which is defined as

$$\underline{\nabla} \cdot \mathbf{u} = \operatorname{tr}((\mathbb{I} - \mathbf{v} \otimes \mathbf{v}) \nabla u),$$

where \otimes denotes the vector direct product. Since $\partial \Omega_F(t) \cap \partial \Omega = \emptyset$, i.e., $\partial \Omega_F(t)$ is a closed interface, no boundary condition is needed for Eq. (6).

2.3. Non-dimensional form of the equations

The governing equations are solved in nondimensional form. Let U_{∞} , L, ρ_2 , C_{∞} , be the characteristic values for velocity, length, density and surfactant concentration C. We define the nondimensional density $\rho(\mathbf{x})$ and Reynolds number $Re(\mathbf{x})$ in different parts of the domain Ω as

$$\rho(\mathbf{x}) = \begin{cases} \rho_1/\rho_2 & \text{for } \mathbf{x} \text{ in } \Omega_1(t), \\ 1 & \text{for } \mathbf{x} \text{ in } \Omega_2(t), \end{cases} \quad Re(\mathbf{x}) = \begin{cases} Re_2\mu_2/\mu_1 & \text{for } \mathbf{x} \text{ in } \Omega_1(t), \\ Re_2 & \text{for } \mathbf{x} \text{ in } \Omega_2(t), \end{cases} \quad Re_2 = \frac{\rho_2U_{\infty}L}{\mu_2}.$$

Furthermore, we define the nondimensional variables as

$$\tilde{x} = \frac{x}{L}, \quad \tilde{u} = \frac{u}{U_{\infty}}, \quad \tilde{w} = \frac{w}{U_{\infty}}, \quad \tilde{t} = \frac{tU_{\infty}}{L}, \quad \tilde{p} = \frac{p}{\rho_2 U_{\infty}^2}, \quad \tilde{T} = \frac{\Gamma}{\Gamma_{\infty}}, \quad \tilde{C} = \frac{C}{C_{\infty}}.$$

Using these nondimensional variables and omitting the tilde afterwards, we can write the nondimensional form of the timedependent Navier–Stokes equations (1) as

$$\rho(\mathbf{x})\left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla)\mathbf{u}\right) - \nabla \cdot (\mathbb{S}_k(\mathbf{u}, p)) = \rho(\mathbf{x})\frac{\mathbf{e}}{Fr}, \quad \nabla \cdot \mathbf{u} = \mathbf{0}.$$
(7)

The nondimensional form of the kinematic condition becomes

$$\mathbf{w}\cdot\mathbf{v}=\mathbf{u}\cdot\mathbf{v},\quad [\mathbf{u}]=\mathbf{0}$$

and the force-balancing boundary condition with the linear equation of state (2) can be written as

$$\mathbf{v}[\mathbb{S}(u,p)] \cdot \mathbf{v} = \frac{1}{We} \left(1 + E_1 \left(\frac{\Gamma_1}{\Gamma_\infty} - \Gamma \right) \right) \mathcal{K}, \quad \tau_i[\mathbb{S}(u,p)] \cdot \mathbf{v} = -\frac{E_1}{We} \tau_i \cdot \underline{\nabla} \Gamma, \quad i = 1, 2.$$

Here, E_1 is the surfactant elasticity defined as $E_1 = RT_a \Gamma_{\infty} / \sigma_1$. Similarly, the nondimensional form of force balancing boundary conditions with the nonlinear equation of state (3) can be written as

$$\boldsymbol{v}[\mathbb{S}(\mathbf{u},p)] \cdot \boldsymbol{v} = \frac{1}{We} (1 + E_0 \ln(1-\Gamma))\mathcal{K}, \quad \boldsymbol{\tau}_{i,F}[\mathbb{S}(\mathbf{u},p)] \cdot \boldsymbol{v} = -\frac{E_0}{We} \tau_i \cdot \left(\frac{\nabla\Gamma}{1-\Gamma}\right),$$

where E_0 is the surfactant elasticity defined as $E_0 = RT_a\Gamma_{\infty}/\sigma_0$. Finally, the nondimensional form of the stress tensor $\mathbb{S}_k(\mathbf{u}, p)$ and the nondimensional Weber and Froude numbers are given by

$$\mathbb{S}_k(\mathbf{u},p) := \frac{2}{Re(\mathbf{x})} \mathbb{D}(\mathbf{u}) - p\mathbb{I}, \quad We = \frac{\rho_2 U_\infty^2 L}{\sigma_1}, \quad Fr = \frac{U_\infty^2}{Lg}.$$

Note that σ_0 instead of σ_1 has to be used in the definition of the Weber number when the nonlinear equation of state (3) is used.

Remark 1. Often the characteristic velocity in interface flows is defined as $U_{\infty} = \sqrt{Lg}$. In this case the Froude number reduces to 1 and the Weber number becomes the Eötvös number

$$E\mathbf{o} = \frac{\rho_2 \mathbf{g} \mathbf{L}^2}{\sigma_1}.$$

Next, using these nondimensional variables in Eq. (4), and omitting the tilde afterwards, the nondimensional form of the surfactant concentration equation in the outer phase is given by

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C = \nabla \cdot \left(\frac{1}{Pe_c} \nabla C\right). \tag{8}$$

The corresponding boundary condition on $\partial \Omega_F(t)$ with the source term (5) becomes

$$\mathbf{v} \cdot \frac{1}{Pe_c} \nabla C = -\beta C (1 - \Gamma) + Bi Da \Gamma.$$

Similarly, using the nondimensional variables in Eq. (6), and omitting the tilde afterwards, we obtain the nondimensional form of the surfactant concentration equation in the interface with the source term (5) as

$$\frac{\partial\Gamma}{\partial t} + U \cdot \underline{\nabla}\Gamma + \Gamma \underline{\nabla} \cdot \mathbf{u} = \underline{\nabla} \cdot \frac{1}{Pe_s} \underline{\nabla}\Gamma + \frac{\beta}{Da} C(1 - \Gamma) - Bi\Gamma.$$
(9)

Here, the nondimensional numbers (Peclet, Biot, Damköhler and β) in Eqs. (8) and (9) are given by

$$Pe_c = \frac{U_{\infty}L}{D_c}, \quad Pe_s = \frac{U_{\infty}L}{D_s}, \quad Bi = \frac{K_dL}{U_{\infty}}, \quad Da = \frac{\Gamma_{\infty}}{LC_{\infty}}, \quad \beta = \frac{K_a\Gamma_{\infty}}{U_{\infty}}.$$

3. Numerical solution procedure

3.1. ALE approach for time-dependent domains

In the model problem considered, the boundary of Ω is fixed over time but the interface $\partial \Omega_F(t)$ between the two sub-domains $\Omega_k(t)$, k = 1, 2, has to be tracked. For this, we use, as in [14] for free surface flows, the arbitrary Lagrangian–Eulerian (ALE) approach. The domain Ω is decomposed in such a way that the interface $\partial \Omega_F(t)$ is represented in each time step by certain faces of mesh cells. The collection of mesh cells belonging to Ω_1 will be called the inner mesh, those in Ω_2 the outer mesh. On this interface-resolving inner and outer mesh we discretize the time-dependent Navier–Stokes equation (7) and the surfactant concentration Eq. (8) in space. Note that we do not need a separate lower-dimensional mesh to discretize the interface surfactant concentration Eq. (9) in space, we use directly the discrete representation of the interface $\partial \Omega_F(t)$.

In order to find the new shape of the subdomains Ω_1 and Ω_2 for the next time step $t = t^n$, we solve first the time-dependent Navier–Stokes equations (7) with the surfactant concentration Γ at the interface from the previous time step $t = t^{n-1}$, and then solve Eqs. (8) and (9). We move the interface with the fluid velocity \mathbf{u}^n , see Fig. 1, left. Let Ψ_F^n be the resulting displacements of the nodes at the interface. Then, we compute the displacements Ψ_1^n and Ψ_2^n of nodes inside both phases by solving the linear elasticity problem (elastic mesh update [13,20])

$$\nabla \cdot \mathbb{T}(\Psi_k^n) = 0 \quad \text{in } \Omega_k(t^{n-1}), \quad \Psi_k^n = \Psi_k^n \quad \text{on } \partial \Omega_k(t^{n-1}), \quad \Psi_2^n = 0 \quad \text{on } \partial \Omega$$
(10)

for k = 1, 2. Here, T denotes the stress tensor given by

$$\mathbb{T}(\phi) = \lambda_1 (\nabla \cdot \phi) \mathbb{I} + 2\lambda_2 \mathbb{D}(\phi),$$

where λ_1 and λ_2 are the Lame constants (chosen to be $\lambda_1 = \lambda_2 = 1$ in our numerical tests). From the displacement vectors Ψ_k^n , k = 1, 2, we compute the mesh velocity $\mathbf{w}_k^n \approx \Psi_k/(t^n - t^{n-1})$ at time $t = t^n$ in Ω_k , k = 1, 2, which appears in the ALE form of the time-dependent Navier–Stokes equations (7):

$$\rho(\mathbf{x})\left(\frac{\partial \mathbf{u}}{\partial t} + ((\mathbf{u} - \mathbf{w}) \cdot \nabla)\mathbf{u}\right) - \nabla \cdot (\mathbb{S}_k(\mathbf{u}, p)) = \rho(\mathbf{x})\frac{\mathbf{e}}{Fr}, \quad \nabla \cdot \mathbf{u} = \mathbf{0}.$$
(11)

Similarly, the ALE formulation of the surfactant concentration equation in the outer phase reads:

$$\frac{\partial C}{\partial t} + (\mathbf{u} - \mathbf{w}) \cdot \nabla C = \nabla \cdot \left(\frac{1}{Pe_c} \nabla C\right). \tag{12}$$

Since we move the interface with the fluid velocity in the ALE approach, the domain $\partial \Omega_F(t)$ for the surface concentration Eq. (9) is treated in a Lagrangian manner. As a consequence, the Lagrangian form of Eq. (9) will be used:

$$\frac{\partial \Gamma}{\partial t} + \Gamma \underline{\nabla} \cdot \mathbf{u} = \underline{\nabla} \cdot \frac{1}{Pe_s} \underline{\nabla} \Gamma + \beta_{\Gamma} C(1 - \Gamma) - Bi\Gamma.$$
(13)



Fig. 1. Mesh handling. Step 1: move the interface points with the velocity to the new position (left). Step 2: compute the displacements of inner points by an elastic mesh update (right).

Occasionally, the distortion of the cells may become very large after several time steps. In such situations, we remesh the domain and interpolate the solutions from the old mesh to the new mesh. In general, the interpolated fluid velocity will not be discretely divergence free on the new mesh. Therefore, an additional fixed point iteration step of the Navier-Stokes solver with the interpolated velocity as an initial guess is performed before continuing the time stepping scheme.

3.2. Variational form

The finite-element method for solving the governing equations (11)-(13) of the two-phase flows with soluble surfactants is based on a variational form. Let $V := (H_0^1(\Omega))^3$ and $Q := L_0^2(\Omega)$ be the usual Sobolev spaces. We multiply the momentum and mass balance equations (11) by test functions $\mathbf{v} \in V$ and $q \in Q$, respectively, integrate over Ω , and integrate by parts in each sub-domain $\Omega_k(t)$, k = 1, 2, separately. Incorporating the boundary conditions, we get the variational form of (11):

For given Ω_0 , \mathbf{u}_0 , and \mathbf{w} , find $(\mathbf{u}, p) \in V \times Q$ such that

$$\left(\rho(\mathbf{x})\frac{\partial \mathbf{u}}{\partial t}, \mathbf{v}\right) + a((\mathbf{u} - \mathbf{w}), \mathbf{u}, \mathbf{v}) - b(p, \mathbf{v}) + b(q, \mathbf{u}) = (f, \mathbf{v}), \quad \forall (\mathbf{v}, q) \in V \times Q.$$
(14)

Here, (\cdot, \cdot) denotes the inner product in $L^2(\Omega)$ and its vector-valued versions. The bilinear forms $a(\hat{\mathbf{u}}, \cdot, \cdot)$ and $b(\cdot, \cdot)$ are given by

$$\begin{split} a(\hat{\mathbf{u}},\mathbf{u},\mathbf{v}) &= 2 \int_{\Omega} \frac{1}{Re(\mathbf{x})} \mathbb{D}(\mathbf{u}) : \mathbb{D}(\mathbf{v}) d\mathbf{x} + \int_{\Omega} \rho(\mathbf{x}) (\hat{\mathbf{u}} \cdot \nabla) \mathbf{u} \cdot \mathbf{v} d\mathbf{x}, \\ b(q,\mathbf{v}) &= \int_{\Omega} q \nabla \cdot \mathbf{v} d\mathbf{x}. \end{split}$$

Furthermore, for the linear equation of state (2), we have

$$(f,\mathbf{v}) = \frac{1}{Fr} \int_{\Omega} \rho(\mathbf{x}) \mathbf{e} \cdot \mathbf{v} d\mathbf{x} - \frac{1}{We} \int_{\partial \Omega_{F}(t)} \left(1 + E_1 \left(\frac{\Gamma_1}{\Gamma_{\infty}} - \Gamma \right) \right) (\mathbf{v} \cdot \mathbf{v}) \mathcal{K} dS - \frac{E_1}{We} \int_{\partial \Omega_{F}(t)} (\mathbf{v} \cdot \tau_i) (\underline{\nabla} \Gamma) \cdot \tau_i dS$$
(15)

and for the nonlinear equation of state (3), we have

$$(f, \mathbf{v}) = \frac{1}{Fr} \int_{\Omega} \rho(\mathbf{x}) \mathbf{e} \cdot \mathbf{v} d\mathbf{x} - \frac{1}{We} \int_{\partial \Omega_{F}(t)} (1 + E_0 \ln(1 - \Gamma)) (\mathbf{v} \cdot \mathbf{v}) \mathcal{K} dS - \frac{E_0}{We} \int_{\partial \Omega_{F}(t)} (\mathbf{v} \cdot \tau_i) \left(\frac{\nabla \Gamma}{1 - \Gamma}\right) \cdot \tau_i dS.$$
(16)

The curvature \mathcal{K} in the surface integrals of Eqs. (15) and (16), respectively is replaced by the Laplace–Beltrami operator Δ of the identity *id*, more precisely, $\mathcal{K}v = -\underline{\Delta}id$. Then, applying integration by parts we reduce one order of differentiation associated with the curvature. Hence, the surface integrals in Eqs. (15) and (16), which contain the curvature \mathcal{K} , become

$$-\frac{1}{We}\int_{\partial\Omega_{F}(t)}\underline{\nabla}id:\underline{\nabla}\Big(\mathbf{v}\Big[1+E_{1}\Big(\frac{\Gamma_{1}}{\Gamma_{\infty}}-\Gamma\Big)\Big]\Big)dS=-\frac{1}{We}\int_{\partial\Omega_{F}(t)}\underline{\nabla}id:\Big(\Big[1+E_{1}\Big(\frac{\Gamma_{1}}{\Gamma_{\infty}}-\Gamma\Big)\Big]\underline{\nabla}\mathbf{v}-E_{1}\underline{\nabla}\Gamma\otimes\mathbf{v}\Big)dS$$

and

$$-\frac{1}{We}\int_{\partial\Omega_{F}(t)}\underline{\nabla}id:\underline{\nabla}(\mathbf{v}[1+E_{0}\ln(1-\Gamma)])dS=-\frac{1}{We}\int_{\partial\Omega_{F}(t)}\underline{\nabla}id:\left([1+E_{0}\ln(1-\Gamma)]\underline{\nabla}\mathbf{v}-\left[\frac{E_{0}}{1-\Gamma}\underline{\nabla}\Gamma\right]\otimes\mathbf{v}\right)dS,$$

respectively.

Next, for deriving a variational form of the soluble surfactant concentration Eqs. (8) and (9), we define $G := H^1(\Omega_2(t))$ and $M := H^1(\partial_F \Omega(t))$. Multiplying Eqs. (8) and (9) by test functions $\phi \in G$ and $\psi \in M$, integrating over $\Omega_2(t)$ and $\partial_F \Omega(t)$, respectively, incorporating the boundary conditions for C, we obtain the coupled problem for the soluble surfactant concentration: For given $(C_0, \Gamma_0, \mathbf{w})$, find $(C, \Gamma) \in G \times M$ such that for all $(\phi, \psi) \in G \times M$

$$\left(\frac{\partial C}{\partial t},\phi\right)_{\Omega_{2}(t)} + a_{c}((\mathbf{u}-\mathbf{w}),C,\phi) + b_{c}(C,\Gamma,\phi) = s_{c}(\Gamma,\phi),$$

$$\left(\frac{\partial \Gamma}{\partial t},\psi\right)_{\partial\Omega_{F}(t)} + a_{\Gamma}(\mathbf{u},\Gamma,\psi) + b_{\Gamma}(\Gamma,C,\psi) = s_{\Gamma}(C,\psi),$$
(17)
(18)

where

$$\begin{aligned} a_{c}(\mathbf{u},C,\phi) &= \frac{1}{Pe_{c}} \int_{\Omega_{2}(t)} \nabla C \cdot \nabla \phi d\mathbf{x} + \int_{\Omega_{2}(t)} (\mathbf{u} \cdot \nabla) C \phi d\mathbf{x}, \\ b_{c}(\Gamma,C,\phi) &= \beta \int_{\partial \Omega_{F}(t)} (1-\Gamma) C \phi dS, \\ s_{c}(\Gamma,\phi) &= BiDa \int_{\partial \Omega_{F}(t)} \Gamma \phi dS, \end{aligned}$$

$$\begin{split} a_{\Gamma}(\mathbf{u},\Gamma,\psi) &= \frac{1}{Pe_s} \int_{\partial\Omega_{F}(t)} \underline{\nabla}\Gamma \cdot \underline{\nabla}\psi dS + \int_{\partial\Omega_{F}(t)} \Gamma \underline{\nabla} \cdot \mathbf{u}\psi dS, \\ b_{\Gamma}(C,\Gamma,\psi) &= \frac{\beta}{Da} \int_{\partial\Omega_{F}(t)} C\Gamma\psi dS + Bi \int_{\partial\Omega_{F}(t)} \Gamma\psi dS, \\ s_{\Gamma}(C,\phi) &= \frac{\beta}{Da} \int_{\partial\Omega_{F}(t)} C\psi dS. \end{split}$$

3.3. Spatial and temporal discretization

Let $0 = t^0 < t^1 < \cdots < t^N = T$ be a decomposition of the considered time interval [0,T] and $\delta t = t^n - t^{n-1}$ the time step when going from t^{n-1} to t^n . Then, the time derivative is discretized by the fractional-step- θ scheme [31, Chapter 3.2.1] which is – on fixed domains – strongly A-stable and of second-order convergent.

The choice of an appropriate discretization in space for the Navier–Stokes equations depends on several aspects. In our numerical computations, we consider a 3D-axisymmetric configuration, which allows two-dimensional finite elements for the quantities on the cross-section and one-dimensional elements for approximating Γ . Since first derivatives of the fluid velocity are present in the surfactant transport Eq. (18), second-order approximations for the fluid velocity are advisable. Next we are interested in inf-sup stable discretizations [16] for velocity and pressure. Furthermore, we prefer discontinuous pressure approximations which guarantee the local mass conservation of the fluid. From the approximation point of view, one possibility would be the Scott–Vogelius finite-element pair (P_2, P_1^{disc}) , i.e., continuous piecewise quadratic polynomials for the velocity components and discontinuous piecewise linears for the pressure. Unfortunately, this pair is not inf-sup stable on general shape-regular meshes and it seems hopeless to try to satisfy the additional mesh constraints that guarantee stability on moving meshes. Therefore, we propose the inf-sup stable finite-element pair $(P_2^{bubble}, P_1^{disc})$, i.e., continuous piecewise quadratic polynomials enriched by a cubic bubble function for the velocity components and discontinuous piecewise linears for the pressure [16]. For the spatial discretization of the surfactant concentration in the bulk and on the interface, continuous piecewise guadratic finite elements are used. Suppressing spurious velocities is an important challenge in the computation of two-phase flows. As shown in [12], errors in the approximation of the boundary and its curvature as well as in the discontinuous pressure are the main causes for the generation of spurious velocities in finite-element methods. We suppress spurious velocities by using an interface-resolving mesh, a discontinuous pressure approximation, and isoparametric finite elements close to the interface for a better approximation of curved boundaries [12]. For the elastic mesh update we need only the position of vertices in the mesh. Thus, continuous piecewise linear finite elements are sufficient to approximate the displacement components in Eq. (10). Fig. 2 gives an overview of the finite elements used.

3.4. Linearization of the nonlinear discrete systems

The nonlinear convection term in the Navier–Stokes equations (14) is handled by a fixed point iteration as in [13]. At time t^n , starting with $\mathbf{u}_0^n := \mathbf{u}^{n-1}$, $\mathbf{w}_0^n := \mathbf{w}^{n-1}$ and replacing the form $a(\mathbf{u}^n - \mathbf{w}^n, \mathbf{u}^n, \mathbf{v})$ by $a(\mathbf{u}_{i-1}^n - \mathbf{w}_{i-1}^n, \mathbf{u}_i^n, \mathbf{v})$, i = 1, 2, ..., we iterate until the residual of the Navier–Stokes equations becomes less than 10^{-8} . Note that in each step of the fixed point iteration, the linear elasticity problem has to be solved to determine the mesh velocity \mathbf{w}_i^n , i = 1, 2, ..., 'virtually' without moving the mesh.

After stopping the fixed point iteration, we first solve the surfactant Eqs. (17) and (18) before moving the mesh to a new position. For the linearization of the coupled Eqs. (17) and (18) a Gauss–Seidel type fixed point iteration has been used. For instance, with the backward Euler time differencing scheme, the semi-discrete (in time) linearized equations (17) and (18) at time t^n read:

Let $\Gamma_0^n := \Gamma^{n-1}$, for i = 1, 2, ... compute (C_i^n, Γ_i^n) from

$$(C_{i}^{n},\phi) + \delta t \left[a_{c} \left((\mathbf{u}^{n} - \mathbf{w}^{n}), C_{i}^{n},\phi \right) + b_{c} \left(C_{i}^{n}, \Gamma_{i-1}^{n},\phi \right) \right] = \left(C^{n-1},\phi \right) + \delta t s_{c} \left(\Gamma_{i-1}^{n},\phi \right)$$

$$(\Gamma_{i}^{n},\psi) + \delta t \left[a_{\Gamma} \left(\mathbf{u}^{n}, \Gamma_{i}^{n},\psi \right) + b_{\Gamma} \left(\Gamma_{i}^{n}, C_{i}^{n},\psi \right) \right] = (\Gamma^{n-1},\psi) + \delta t s_{\Gamma} \left(C_{i}^{n},\psi \right).$$

$$(19)$$

In our computations, we stop this iteration when the residual of the first equation becomes less than 10^{-12} . Our numerical study shows that in general this condition is fulfilled within 2 to 4 iteration steps. Also, the computational cost for the ALE



Fig. 2. Finite elements for approximating each component of the velocity, pressure, surfactant concentration in the bulk and on the interface, and displacements of nodes in the elastic mesh update.

elastic mesh tracking (computation of the mesh velocity in all fixed point iteration steps and the mesh movement) is around 2% of the entire computational cost.

4. Numerical experiments

To validate the proposed numerical scheme a number of numerical tests are performed in 3D-axisymmetric configurations. The numerical results are compared with analytical and experimental values. Since the proposed numerical scheme has already been validated in [14] for free surface flows and two-phase flows with insoluble surfactants, we mainly consider here tests related to soluble surfactants. First, a simple test is designed to validate the numerical approximation of the convection terms in the surface concentration equation. Then, an analytical approximation for the diffusion of the bulk concentration including surfactant transport from the bulk phase to the interface is compared with the numerical solution. Finally, an array of computations for a rising bubble problem with and without soluble surfactants are performed.

4.1. Convection test (insoluble)

We consider a unit sphere which is continuously expanding in the normal direction with the prescribed divergence-free velocity

$$\mathbf{u} = \frac{1}{R^3} (x(t), y(t), z(t)), \quad R(t) = (x^2(t) + y^2(t) + z^2(t))^{1/2}, \quad \frac{d\mathbf{x}}{dt} = \mathbf{u}.$$

We assume that there is no diffusion on the interface, i.e., $D_s = 0$, the initial surfactant concentration on the interface is uniform, and there is no mass transfer between the bulk phase and the interface, i.e., $k_a = k_d = 0$. For this configuration, the surfactant concentration on the interface changes solely due to the change in the surface area of the sphere. Since the mass of the surfactant is constant over time, we have $\Gamma(t) = 1/R(t)^2$ for an initial surfactant concentration $\Gamma_0 = 1$, and the mass loss

$$\omega(t):=4\pi-\int_{\partial\Omega_F(t)}\Gamma(t)ds$$

is zero.

In the numerical computation, we started with a coarse mesh of 51 degrees of freedom for the surfactant concentration and generated the finer meshes by uniform refinements (from mesh size h = 1/8 for level 1 to mesh size h = 1/128 for level 5). Furthermore, in order to keep the temporal discretization error small enough, a very small time step $\delta t = 6.25 \times 10^{-5}$ has been used. The mean value of the obtained numerical solution $\Gamma_h(t)$ on different mesh levels 1, 3, 5 and the analytical solution $\Gamma(t)$ are plotted in Fig. 3 (left). The numerical and the analytical solutions are in excellent agreement. Due to the scaling in Fig. 3 (left), differences in the numerical solutions obtained on different mesh levels are not visible. Therefore, we computed also the relative error in the mean value of $\Gamma_h(t)$

rel. errorin
$$\Gamma_{mean} := 1 - \frac{\int_{\partial \Omega_F(t)} \Gamma_h(t) ds}{\int_{\partial \Omega_F(t)} \Gamma(t) ds}.$$

The relative error in Γ_{mean} for all mesh levels are plotted in Fig. 3 (right), and the mesh convergence behaviour can be seen clearly. The mass loss $\omega_h(t)$ in the surfactant concentration over a period of time has been computed for different mesh levels with respect to the L^{∞} and L^2 norm as follows



Fig. 3. Numerical and analytical solutions for the change in the surfactant concentration on the interface (left) and relative error (right) over time for different mesh levels in a continuously expanding sphere test case.

$$\|\text{mass loss in}\Gamma\|_{\infty} = \max_{(0,T)} |\omega_h(t)|, \quad \|\text{mass loss in }\Gamma\|_0 = \left(\int_0^T |\omega_h(t)|^2 dt\right)^{1/2}.$$

The results for T = 2 are shown in Fig. 4. Even on the coarsest mesh, the mass loss is less than one percent and reduces further with order 2 when refining the mesh. These results demonstrate the high accuracy and the convergence of the numerical scheme.

4.2. Bulk concentration diffusion and mass transfer test

In this test case we verify the diffusion of the bulk concentration and the mass transfer from the bulk phase to the interface. A similar test has been performed for the front-tracking method in [24]. We consider a sphere with radius $r_0 = 1$ suspended in a large cylinder. We assume that the interface is initially clean, i.e. $\Gamma_0 = 0$, and that the initial bulk concentration $C_0 = 1$. Furthermore, we assume that the mass transfer is solely due to the molecular diffusion. Moreover, a simplified version of the source term (5) as $S(\Gamma, C) = k_aC$ is considered, so that the mass transfer takes place always from the bulk phase to the interface. For a short period of time or in an infinite domain, an analytical approximation of the dimensionless bulk concentration C is given by

$$C = 1 - \frac{k_a \sqrt{\pi D_c t} / D_c}{1 + \sqrt{\pi D_c t} (1 + k_a r_0 / D_c) / r_0} \frac{r_0}{r} \operatorname{erfc}\left(\frac{r - r_0}{2\sqrt{D_c t}}\right),$$
(20)

where $\operatorname{erfc}(x)$ is the complementary error function. Using (20), the analytical approximation of the dimensionless interface concentration is given by

$$\Gamma = \Gamma_0 + k_a C_\infty \left(t - \frac{a\omega}{\eta^3} \left(\eta^2 t - 2\eta\sqrt{t} + 2\ln\left(1 + \eta\sqrt{t}\right) \right) \right),\tag{21}$$

where $\omega = k_a/D_c$, $a = \sqrt{\pi D_c}$ and $\eta = a(1 + r_0 \omega)/r_0$. The droplet is placed at the center of the cylindrical tube that extends five and eight droplet radii in the radial (*r*-) and axial (*z*-) directions, respectively. In our computation, an unstructured grid has been used with 401 and 8804 degrees of freedom for Γ and *C*, respectively. Furthermore, we used $k_a = 1$, $D_c = 0.1$ and $C_{\infty} = 1$. For illustrating the transport of the bulk concentration to the interface, contour plots of the bulk surfactant concentration at various times are shown in Fig. 5. As expected, the contours of *C* are symmetric which validates qualitatively the computational results. For a quantitative comparison with the analytical approximation (20), the bulk concentration profiles taken along the horizontal plane through the center of the droplet are presented in Fig. 6 (left) at different times. The computational results are in good agreement with the analytical approximation. Finally, to validate the mass transfer from the bulk phase to the interface, we compare the computational results for the interface concentration with the analytical approximation (21) in Fig. 6 (right) over a period of time. The figure clearly shows that the numerical solution agrees well with the analytical approximation. Furthermore, as in the previous example, we performed a mesh convergence test and observed that the numerical solution converges to the analytical approximation when the mesh is refined. In Fig. 6 (right), we skip numerical results obtained on different grids since there are no visible differences as in the previous example.

4.3. Rising bubble with surfactants

In the next section, we first compare a few quantities such as the terminal velocity and the sphericity of a clean bubble with the experimental observations reported in [10]. Then, for the same test example (which will be called the experimental bubble configuration in the following), we study the influence of surfactants on the dynamics of the rising bubble for



Fig. 4. Surfactant mass losses measured in L^2 and L^∞ norms over time for different mesh levels in a continuously expanding sphere test case.

S. Ganesan, L. Tobiska/Journal of Computational Physics 231 (2012) 3685-3702



Fig. 5. Bulk concentration diffusion. Contours of the bulk surfactant concentration at dimensionless times t = 1, 5, 8, and 10.



Fig. 6. Bulk concentration diffusion (left) and mass transfer test (right). Comparison of numerical and analytical approximation of the bulk concentration profiles taken along the horizontal plane through the center of the droplet at dimensionless times t = 0.1, 1, 4, and 7 (left). Comparison of numerical and analytical interface concentration over a period of time (right).

different values of Γ_0 , *Bi* and *Pe_c*. In the final test example, we provide a set of reference values for the benchmark rising bubble (test case 1 in [18]) with soluble surfactants in a 3D-axisymmetric configuration.

In both, the experimental and the benchmark bubble configurations, we consider a rising bubble driven by buoyancy force in quiescent water. In our model, Ω_1 and Ω_2 denote the air and water column, respectively. Furthermore, in all computations of the rising bubble problem with surfactants we used an uniform initial concentration $C_0 = 1$ in the bulk phase. For the experimental bubble, the following material parameters are used: the density $\rho_1 = 1.23 \text{ kg/m}^3$, $\rho_2 = 1000 \text{ kg/m}^3$, the dynamic viscosity $\mu_1 = 1.73 \times 10^{-5} \text{ N s/m}^2$, $\mu_2 = 1 \times 10^{-3} \text{ N s/m}^2$, the surface tension 0.073 N/m and the gravitational constant $g = 9.8 \text{ m/s}^2$. It is assumed that the initial shape of the bubble is spherical with the diameter $d_0 = 2.5 \times 10^{-3} \text{ m}$, and the bubble is at rest initially, i.e. $\mathbf{u}(0, \mathbf{x}) = \mathbf{0}$. The computational domain Ω , which contains the bubble Ω_1 , is a cylindrical vessel with diameter $2 \times 10^{-2} \text{ m}$ and height $2 \times 10^{-1} \text{ m}$, respectively. For the benchmark bubble, the following material parameters are used: the density $\rho_1 = 100 \text{ kg/m}^3$, $\rho_2 = 1000 \text{ kg/m}^3$, the dynamic viscosity $\mu_1 = 1 \text{ N s/m}^2$, $\mu_2 = 10 \text{ N s/m}^2$, the surface tension 24.5 N/m and a reduced gravitational constant $g = 0.98 \text{ m/s}^2$. Furthermore, it is assumed that the initial shape of the bubble is at rest initially. The resulting dimensionless numbers

Table 1			
Dimensionless numbers for	both the experimental	and benchmark	bubble examples.

	Re ₂	Ео	Fr	$ ho_2/ ho_1$	μ_2/μ_1	Pec	Pes	Da
Experimental	1107	3.36	1	813	57.8	1	10	200
Benchmark	99	40	1	10	10	1	10	200

obtained by choosing the characteristic values $L = 2d_0$ and $U_{\infty} = \sqrt{Lg}$ for both the experimental and benchmark bubbles, are given in Table 1. Furthermore, the behaviour of the LEOS Eq. (2) and NLEOS Eq. (3) for the test examples considered with $E_0 = E_1 = 0.5$ and $\Gamma_1 = 0$ are depicted in Fig. 7. It can be seen clearly that the LEOS and the NLEOS behave almost identically when the surfactant concentration is low. Thus, the effects of both the LEOS and the NLEOS on the bubble dynamics can be expected to be very similar, and this is observed in our numerical computations. Therefore, for both the experimental and the benchmark test example, we present the numerical results only for the LEOS.



Fig. 7. Normalized surface tension in dependence of the surfactant concentration for a linear (LEOS) and nonlinear (NLEOS) equation of state. The case $E_0 = E_1 = 0.5$ and $\Gamma_1 = 0$.



Fig. 8. Influence of Γ_0 : The rise velocity, the kinetic energy, the center of mass and the sphericity of the experimental bubble for different Γ_0 cases.

In order to study the influence of the surfactants on the dynamics of the rising bubble, we compute the rise velocity, the kinetic energy, the center of mass (z variable) and the sphericity of the bubble. The velocity component directed opposite and parallel to the gravitational force is referred to as the rise velocity. The rise velocity and the kinetic energy are computed by

rise velocity =
$$\frac{1}{|\Omega_1(t)|} \int_{\Omega_1(t)} u_z r \, dr \, dz$$
, kinetic energy = $\frac{1}{|\Omega_1(t)|} \int_{\Omega_1(t)} \mathbf{u} \cdot \mathbf{u} r \, dr \, dz$,

where $|\Omega_1(t)| = \int_{\Omega_1(t)} r \, dr \, dz$. If the rise velocity reaches a stationary value for $t \to \infty$, then the stationary value is called the terminal velocity. The center of mass in the *z* component and the sphericity of the bubble are computed by

center of mass
$$=\frac{1}{|\Omega_1(t)|}\int_{\Omega_1(t)} zr dr dz$$
, sphericity $=\frac{\text{surface area of volume-equivalent sphere}}{\text{surface area of the bubble}}$

This implies that the sphericity of a spherical bubble will be 1 and for any other deformed bubble it will be less than 1.

4.4. Experimental bubble

4.4.1. Influence of Γ_0

First, to study the influence of the initial surface surfactant concentration Γ_0 on the dynamics of the rising bubble we consider the following four cases: (i) clean bubble, (ii) Case A: $\Gamma_0 = 0$, (iii) Case B: $\Gamma_0 = 0.2$, and (iv) Case C: $\Gamma_0 = 0.5$. In all three surfactant cases, we used Bi = 1 and $\beta = 200$. The numerically computed rise velocity, the kinetic energy, the center of mass and the sphericity of the bubble are presented in Fig. 8. For the clean bubble, the dimensionless rise velocity reaches a stationary value (terminal velocity) of 1.4 (approximately), which is equivalent to 3.09×10^{-1} m/s. Furthermore, it is observed that the shape of the spherical bubble deforms to an elliptic shape during this transition. In experiments, it has been reported that the terminal velocity of 3×10^{-1} m/s (approximately) and the shape of the bubble belongs to the ellipsoidal regime for the clean bubble data [10]. In the Case A, the bubble is clean initially, and thus the rise velocity is initially similar to the clean



Fig. 9. Position and the flow dynamics of the rising bubble at dimensionless times t = 0.5, 1.0, 1.5, and 2.0, in the case A (top row) and case C (bottom row). At t = 2.0, the position of the bubble in the case A is approximately z = 2.4, whereas the position of the bubble in the case C is approximately z = 0.9. The arrows represent the flow direction and the colours represent the magnitude of the fluid velocity.

bubble case. However, a retarding effect on the rising bubble is observed when the surfactant concentration Γ increases gradually due to the domination of adsorption over desorption. In the Cases B and C, initially the rise velocity increases similar to the clean bubble. However, after a very short period of time, the rise velocities in both cases decrease considerably and reach stationary values quickly. In all cases, the kinetic energies of the bubbles behave similarly to their corresponding rise velocity. The center of mass of the bubble indicates how far the bubble rose. The interesting observations are related to the Cases B and C, in which the bubble rose significantly slow. The surfactant tends to immobilize the surface and reduce the slip (or tangential) velocity, and slows the bubbles rise by increasing the viscous drag on it. For insoluble, diffusion-free surfactant in the steady state, parts of an interface that are covered by surfactant have zero slip velocity. Furthermore, the bubble remains spherical when the surface surfactant concentration is high. In Fig. 9 the positions reached and the different shapes of the bubbles at dimensionless times t = 0.5, 1.0, 1.5, and 2.0, for the Cases A and C are shown.

4.4.2. Influence of Bi

In order to study the influence of *Bi* on the dynamics of a rising bubble, we fix the values of β and the initial surface concentration Γ_0 as 100 and 0.2, respectively. For these data, we performed an array of computations with (i) clean bubble, (ii) Case D : *Bi* = 0, (iii) Case E: *Bi* = 1, and (iv) Case F: *Bi* = 10. The rise velocity, the kinetic energy, the center of mass and the sphericity of the bubble obtained from the numerical computations are presented in Fig. 10. For the Cases D and E, the rise velocities lead to stationary values very quickly due to high surface surfactant concentration. When *Bi* is large, the surfactant leaves the interface quickly and it accelerates the rise velocity. A similar effect is observed on the kinetic energy of the rising bubble and it shows that the rise velocity dominates the radial velocity of the bubble. It is clear from the behavior of rise velocity that the rate of change in the center of mass with respect to time increases when *Bi* increases. The sphericity of the bubble depends on the acceleration of the rise velocity, i.e., the bubbles keep their sphericity when the acceleration of the rise velocity is low.

4.4.3. Influence of bulk Peclet number (Pe_c)

In the previous computations of contaminated rising bubbles with $Pe_c = 1$, the value of bulk diffusivity $D_c = 1.1 \times 10^{-3} \text{ m}^2/\text{s}$ is used. However, many surfactants consist of large molecules that have low bulk diffusivity, i.e., high Pe_c . In such cases, a narrow transition layer in which the concentration of the bulk surfactants varies rapidly will appear close to the interface. As mentioned in the introduction, spurious oscillations in the numerical solution can be avoided by choosing the mesh size (at least near the interface) small enough. We considered three test cases, $Pe_c = 1$, $Pe_c = 10$ and $Pe_c = 1000$ and used very fine meshes close to the interface. The values of the remaining parameters are chosen from the previous test Case F, see Section 4.4.2. As expected no oscillations, even in the case $Pe_c = 1000$, have been observed in our computations. The surface surfactant concentration (Γ) along the interface at different times is depicted in Fig. 11. The value of Γ along the entire interface is



Fig. 10. Influence of Bi: the rise velocity, the kinetic energy, the center of mass and the sphericity of the experimental bubble for different Bi cases.

smaller than the values of Γ in the other two cases. Furthermore, for $Pe_c = 1$ it is nearly zero at the top of the bubble. The curve for t = 0.125 intersects the curve for t = 0.5 which is cut by the curve for t = 1. The increase in surface surfactant concentration toward the rear stagnation point (arclength = 0) can be explained by the advection of surfactant along the interface while the bubble rises. Later on, the surfactant near the rear part of the bubble starts to leave the interface into the bulk phase by desorption when a sufficiently high surface concentration develops. This can also be seen in Fig. 12, where the



Fig. 11. Surface surfactants concentration (Γ) profile along the interface (bottom to top) at different instances for the experimental bubble test case with $\Gamma_0 = 0.2$ and $Pe_c = 1$ (left), $Pe_c = 10$ (right), $Pe_c = 1000$ (bottom).



Fig. 12. The contour plots of the bulk surfactant concentration at time t = 4 for the experimental bubble test case with $\Gamma_0 = 0.2$ and $Pe_c = 1$ (left), $Pe_c = 10$ (middle), $Pe_c = 1000$ (right).

contour plots of the bulk surfactant concentration at time t = 4 for the test cases $Pe_c = 1$, $Pe_c = 10$, and $Pe_c = 1000$ are plotted. The bulk surfactant concentration adjacent to the interface, especially, at the bottom of the rising bubble, is higher for low bulk diffusivity cases; see the scales of different pictures in Fig. 12. Therefore, the adsorption dominates the desorption (refer Eq. (5)) and increases Γ when the bulk diffusivity is reduced. It eventually slows the bubble's rise.

4.5. Benchmark bubble

Next, for the benchmark bubble example we consider the following three cases: (i) clean bubble, (ii) Case G: Bi = 1, $\Gamma_0 = 0.2$, and (iii) Case H: Bi = 10, $\Gamma_0 = 0.5$, and use $\beta = 50$ in surfactant cases. The computationally obtained maximum sur-



Fig. 13. Computationally obtained maximum surface surfactant (left) and the mass of the surface surfactant (right) over the period of time in the benchmark bubble test cases.



Fig. 14. The rise velocity, the kinetic energy, the center of mass and the sphericity of the benchmark bubble for different cases.

face surfactant concentration Γ_{max} at any point $x \in \partial \Omega_F(t)$ and the mass of the surface surfactant over the period of time are plotted in Fig. 13. Even though the value of Γ_{max} is high at the initial stage in the Case H, it decreases quickly and becomes less than the value of Γ_{max} in the Case G. A similar effect is observed for the mass of Γ in the Case H. However in the Case G, initially the mass of Γ reduces slightly, while later on it remains almost constant. For these three test cases, the rise velocity, the kinetic energy, the center of mass, and the sphericity obtained from the computations are presented in Fig. 14. The dimensionless terminal velocity of 0.51 (approximately) is observed for the clean bubble case. Here, the shape of the clean bubble changes from spherical to ellipsoidal over the period of time. An interesting effect of variation in *Bi* can be seen in the rise velocity of the bubble. Even though the rise velocity in the Case H is slightly smaller than the clean bubble initially, the large desorption of the surfactant from the interface to the bulk accelerates the rise velocity of the bubble. Since the desorption of the surfactant from the interface is less in the Case G, the rise velocity acceleration effect has not been observed.

4.6. Mass conservation

An important property reflecting the accuracy of a numerical scheme for two-phase flows with surfactants is the conservation of mass. To verify this property for the proposed numerical scheme, we evaluate the mass fluctuation in both the liquid and surfactants. The relative mass fluctuation of the bubble over time is given by

$$\delta\Omega_1(t) = \frac{|\Omega_1(t)| - |\Omega_1(0)|}{|\Omega_1(0)|} \times 100\%, \quad |\Omega_1(t)| = \int_{\Omega_1(t)} r dr dz.$$

The relative fluctuation of the total surfactant mass over time is computed by

$$\delta_{\Gamma}^{\mathsf{C}} = \frac{M(t) - M(0)}{M(0)} \times 100\%, \quad M(t) = \int_{\Omega_{2}(t)} Cr dr dz + Da \int_{\partial \Omega_{F}(t)} \Gamma r ds.$$



Fig. 15. The relative fluctuation of the mass of the bubble and the total surfactants mass occurred in the experimental bubble for different cases.



Fig. 16. The relative fluctuation of the mass of the bubble and the total surfactants mass occurred in the benchmark bubble for different cases.

3701

The relative mass fluctuation of the bubble and the total surfactant mass fluctuation observed in the experimental bubble for the Cases A, B, and C, are presented in Fig. 15. In the high rise velocity Case A, the mass variation in the bubble is less than 0.08% at the dimensionless time t = 4. Furthermore, in all cases the total surfactant mass variation has been less than 0.8% during the time interval considered. Next, the relative mass fluctuation of the bubble and the total surfactant mass fluctuation observed in the benchmark bubble Cases G and H of our computations are presented in Fig. 16. In all benchmark bubble cases, the mass variations in the bubble are around 0.02% at the dimensionless time t = 5. Furthermore, the total surfactant mass variation among all computations of the benchmark bubble cases is less than 0.25% during the time interval considered. In general, the mass fluctuation is more when the rise velocity and the deformation of the bubble are high.

5. Conclusions

An interface-resolving moving mesh finite-element scheme has been proposed for the simulation of 3D-axisymmetric two-phase flows with soluble surfactants. In this scheme, the surface partial differential equation describing the surfactant transport on the interface is treated in a Lagrangian manner, while all other equations are handled with the arbitrary Lagrangian–Eulerian approach. In our model, we considered a formulation of the surface partial differential equation which avoids the approximation of the curvature of the interface. The curvature arising in the force balance at the interface is replaced by the Laplace–Beltrami operator and incorporated as a boundary condition into the weak formulation of the Navier–Stokes equation. Applying integration by parts to the curvature term, the order of differentiation can be reduced by one. This technique allows us to treat the curvature term semi-implicitly, and to approximate the curvature term with continuous finite-element basis functions. The inf-sup stable finite-element pair (P_2^{bubble} , P_1^{disc}), i.e., continuous piecewise polynomials of second order enriched with cubic bubble functions and discontinuous piecewise polynomials of first order, has been used for the spatial discretization of velocity and pressure. This choice of finite-element pair guarantees the mass conservation cell-wise. Even more importantly, the discontinuous pressure approximation together with an interface-resolving mesh suppresses spurious velocities. The mesh update of the moving meshes is realized by solving a linear elasticity equation at each time step on the previous domain as reference domain. An automatic remeshing algorithm has been implemented to remesh the domain when the quality of the mesh become very poor, say if the minimum angle is less than 10° .

The numerical scheme has been validated for a set of simple test examples with known analytical solutions. Furthermore, an array of computations have been performed for a rising bubble with or without insoluble surfactants. The computed rise velocity and the developed shape of a clean rising bubble have been compared with experimental observations and are in good agreement. In our numerical study of the rising bubble with soluble surfactants the following phenomena have been observed: (1) The presence of surfactant retarding the rising bubble. (2) Large desorption of the surfactant from the interface to the bulk phase accelerates the rise velocity. (3) The presence of very high surfactant concentration on the interface dominates the buoyancy force on the bubble and thus the rise velocity becomes almost zero.

Finally, the accuracy of the numerical scheme is demonstrated through the verification of the mass conservation property for both the fluid and the total surfactant mass. Overall the mass variation in the bubble and the total surfactants are 0.08% and 0.8%, respectively during the time interval considered. Thus, the proposed numerical scheme can also be used to compute reference values to verify newly-developed numerical schemes.

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References

- [1] S. Adami, X. Hu, N. Adams, A conservative SPH method for surfactant dynamics, J. Comput. Phys. 229 (5) (2010) 1909–1926.
- [2] S.L. Anna, H.C. Mayer, Microscale tipstreaming in a microfluidic flow focusing device, Phys. Fluids 18 (2006) 121512.
- [3] M.E. Avery, J. Mead, Surface properties in relation to atelectasis and hyaline membrane disease, AMA J. Dis. Child 1959 (1997) 517-523.
- [4] I. Bazhlekov, P. Anderson, H. Meijer, Numerical investigation of the effect of insoluble surfactants on drop deformation and breakup in simple shear flow, J. Colloid Interf. Sci. 298 (2006) 369–394.
- [5] M. Booty, M. Siegel, A hybrid numerical method for interfacial fluid flow with soluble surfactant, J. Comput. Phys. 229 (10) (2010) 3864–3883.
- [6] M.R. Booty, M. Siegel, Steady deformation and tip-streaming of a slender bubble with surfactant in an extensional flow, J. Fluid Mech. 544 (2005) 243-275.
- [7] J.U. Brackbill, D.B. Kothe, C. Zemach, A continuum method for modeling surface tension, J. Comput. Phys. 100 (1992) 335-354.
- [8] A. Brooks, T. Hughes, Streamline upwind/Petrov–Galerkin formulations for convection dominated flows with particular emphasis on the incompressible Navier–Stokes equations, Comput. Methods Appl. Mech. Eng. 32 (1982) 199–259.
- [9] J. Clements, Surface tension of lung extracts, Proc. Soc. Exp. Biol. Med. 95 (1957) 170-172.
- [10] R. Clift, J.R. Grace, M.E. Weber, Bubbles, Drops and Particles, Dover, 2005.
- [11] C.D. Eggleton, T.-M. Tsai, K.J. Stebe, Tip streaming from a drop in the presence of surfactants, Phys. Rev. Lett. 87 (2001) 048302.
- [12] S. Ganesan, G. Matthies, L. Tobiska, On spurious velocities in incompressible flow problems with interfaces, Comput. Methods Appl. Mech. Eng. 196 (7) (2007) 1193–1202.
- [13] S. Ganesan, L. Tobiska, An accurate finite element scheme with moving meshes for computing 3D-axisymmetric interface flows, Int. J. Numer. Methods Fluids 57 (2) (2008) 119–138.
- [14] S. Ganesan, L. Tobiska, A coupled arbitrary Lagrangian-Eulerian and Lagrangian method for computation of free surface flows with insoluble surfactants, J. Comput. Phys. 228 (8) (2009) 2859–2873.
- [15] S. Ganesan, L. Tobiska, Stabilization by local projection for convection-diffusion and incompressible flow problems, J. Sci. Comput. 43 (3) (2010) 326– 342.

- [16] V. Girault, P.-A. Raviart, Finite Element Methods for Navier–Stokes Equations, Springer-Verlag, Berlin–Heidelberg–New York, 1986.
- [17] H. Hamm, H. Fabel, W. Bartsch, The surfactant system of the adult lung: physiology and clinical perspectives, J. Mol. Med. 70 (1992) 637-657.
- [18] S. Hysing, S. Turek, D. Kuzmin, N. Parolini, E. Burman, S. Ganesan, L. Tobiska, Quantitative benchmark computations of two-dimensional bubble dynamics, Int. J. Numer. Methods Fluids 60 (2009) 1259–1288.
- [19] A.J. James, J. Lowengrub, A surfactant-conserving volume-of-fluid method for interfacial flows with insoluble surfactant, J. Comput. Phys. 201 (2004) 685-722.
- [20] A. Johnson, T. Tezduyar, Mesh update strategies in parallel finite element computations of flow problems with moving boundaries and interfaces, Comput. Methods Appl. Mech. Eng. 119 (1-2) (1994) 73-94.
- [21] M.-C. Lai, Y.-H. Tseng, H. Huang, An immersed boundary method for interfacial flows with insoluble surfactant, J. Comput. Phys. 227 (2008) 7279– 7293.
- [22] J. Lee, C. Pozrikidis, Effect of surfactants on the deformation of drops and bubbles in Navier-Stokes flow, Comput. Fluids 35 (2006) 43-60.
- [23] X. Li, C. Pozrikidis, The effect of surfactants on drop deformation and on the rheology of dilute emulsions in Stokes flow, J. Fluid Mech. 341 (1997) 165– 194.
- [24] M. Muradoglu, G. Tryggvason, A front-tracking method for computation of interfacial flows with soluble surfactants, J. Comput. Phys. 227 (2008) 2238– 2262.
- [25] R.E. Pattle, Properties function and origin of the alveolar lining layer, Nature 175 (1955) 1125-1126.
- [26] C. Pozrikidis, A finite-element method for interfacial surfactant transport, with application to the flow-induced deformation of a viscous drop, J. Eng. Math. 49 (2004) 163–180.
- [27] Y.Y. Renardy, M. Renardy, V. Cristini, A new volume-of-fluid formulation for surfactants and simulations of drop deformation under shear at a low viscosity ratio, Eur. J. Mech. B 21 (2002) 49-59.
- [28] H.-G. Roos, M. Stynes, L. Tobiska, Robust numerical methods for singularly perturbed differential equations, in: Convection-Diffusion-Reaction and Flow Problems, SCM, 24, Springer-Verlag, 2008.
- [29] H. Stone, A. Stroock, A. Ajdari, Engineering flows in small devices: microfluidics toward a lab-on-a-chip, Annu. Rev. Fluid Mech. 36 (1) (2004) 381-411.
- [30] H.A. Stone, A simple derivation of the time-dependent convection-diffusion equation for surfactant transport along a deforming interface, Phys. Fluids A 2 (1) (1990) 111-112.
- [31] S. Turek, Efficient Solvers for Incompressible Flow Problems. An Algorithmic and Computational Approach, Springer-Verlag, Berlin, 1999.
- [32] H. Wong, D. Rumschitzki, C. Maldarelli, On the surfactant mass balance at a deforming fluid interface, Phys. Fluids 8 (1996) 3203-3204.
- [33] J.-J. Xu, Z. Li, J. Lowengrub, H. Zhao, A level-set method for interfacial flows with surfactant, J. Comput. Phys. 212 (2006) 590-616.
- [34] J. Zhang, D. Eckmann, P. Ayyaswamy, A front tracking method for a deformable intravascular bubble in a tube with soluble surfactant transport, J. Comput. Phys. 214 (2006) 366–396.